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The Lawrence Livermore National Laboratory has developed an extensive quality assurance program to provide high quality data and assessments in support of the Marshall Islands Dose Assessment and Radioecology Program. Our quality assurance objectives begin with the premise of providing integrated and cost-effective program support (to meet wide-ranging programmatic needs, scientific peer review, litigation defense, and build public confidence) and continue through from design and implementation of large-scale field programs, sampling and sample preparation, radiometric and chemical analyses, documentation of quality assurance/quality control practices, exposure assessments, and dose/risk assessments until publication. The basic structure of our radioassay quality assurance/quality control program can be divided into four essential elements; (1) sample and data integrity control; (2) instrument validation and calibration; (3) method performance testing, validation, development and documentation; and (4) periodic peer review and on-site assessments. While our quality assurance objectives are tailored towards a single research program and the evaluation of major exposure pathways/critical radionuclides pertinent to the Marshall Islands, we have attempted to develop quality assurance practices that are consistent with proposed criteria designed for laboratory accreditation.

Introduction

Bikini and Enewetak Atolls in the northern Marshall Islands were used in the 1950s by the United States for testing of nuclear weapons. During the early years of the test program, knowledge about the long-term consequences of radioactive fallout and transfer of radionuclides through the local food chain was very limited, and this lead to the prolonged relocation of several atoll communities. Under the auspices of the U.S. Department of Energy (DOE), the Health and Ecological Assessment Division at the Lawrence Livermore National Laboratory (LLNL) has been evaluating radiological conditions at the atolls since 1974. We provide Department of Energy, the United States Congress and the Marshall Islands Government with environmental data and radiological assessments, and help atoll communities make informed decisions about possible resettlement options. We have also implemented an extensive research program to develop and evaluate remedial measures to reduce the dose to returning populations. We now have a very good understanding of radiological conditions on the islands and this knowledge could be used to support a major resettlement effort. Our long-term strategic mission is to develop a policy and guidance to ensure the safe resettlement of the atolls.

Addressing the broad scope of the Marshall Islands program has required the design and development of large scale field programs, the evaluation of all possible exposure pathways and critical radionuclides, development of dose and risk based assessment protocols, understanding of the behaviors, cycling, and transport of radionuclides in atoll ecosystems, and evaluation of different remedial actions to help minimize the uptake of ^{137}Cs into the terrestrial food chain.

The Marshall Islands program maintains and operates facilities to process and analyze up to 10,000 environmental samples per year. Typical matrix types include terrestrial food crops and other vegetation, waters, aerosol particulate, marine biota, soils, and sediments. Data management and sample/data integrity control form very important aspects of the Marshall Islands quality assurance program. Customized computer programs and data management tools have been designed for all database recording and management needs. Radiological doses are calculated using the latest methodologies from ICRP (see Robison et al., 1997 & references therein)[1] and generally include radionuclide data for ^{137}Cs, ^{90}Sr, $^{239+240}Pu$, and ^{241}Am in locally grown foods, a diet model for pertinent local food consumption, external gamma exposure calculations, and exposure via inhalation from radionuclide resuspension. We estimate that the ingestion pathway will contribute 90% of the dose to returning residents, mostly through uptake of ^{137}Cs into terrestrial foods such as coconut, *Pandanus*, breadfruit, and papaya[1-2]. Our measurements are used to guide decisions on many different radiological issues in the Marshall Islands ranging from public health and safety, radiological surveillance, resource development to guidance on remedial strategies. Therefore, the ability to maintain a high level of data quality and credibility within the scientific community and the public domain are crucial to the success of the program.

The Marshall Islands Radioassay Quality Assurance Program (RADQAP)

A well defined and implemented quality assurance program provides the structures, policies, guidance and responsibilities to control and verify all planned and systematic operational activities in maintaining a credible and defensible service. It is essential that a level of traceability be established so that measurements can be related to national standards within a specified uncertainty. Here we discuss the Marshall Islands quality assurance/quality control program under four basic levels of activity, namely; (1) sample and data integrity control; (2) instrument validation and calibration; (3) method performance testing, validation, development and documentation; and (4) periodic peer review and on-site assessments.

<u>Sample and Data Integrity Control</u>

Custom designed computer programs and bar coding are used for all data entry, and to help facilitate sample tracking and integrity control. The basic database and data management structures shown in Fig. 1 are linked through a sample identification number (GAMMA_NUM) generated at the time of sample collection. Bar code labels are generally not used for sample tracking in the field but all relevant information is recorded in field notes and computerized. A field log number is generated and this number eventually served as the sample identification (or gamma number, GAMMA_NUM) with extensions that describe the date of collection, island/atoll location and sample type (Fig. 1). A total of four main database structures are used for sample tracking and data management. Presently these databases are running under dBase 5.0 software (from Aston Tate) and linked through customized FORTRAN programs.

The SAMP.DBF database generated at the time of sample collection acts as a master for other databases as well as being automatically updated with information pertaining to the sample and normalization weight (wet/dry ratio). The SAMP.DBF database also includes description fields for any relevant field, location and laboratory notes. Authorized laboratory personnel cue off the main menu to gain access to the SAMP.DBF, LAB.DBF and SAMPTRAK.BDF databases (Figure 1). Routine sample processing operations are performed in separate soil or vegetation laboratory facilities depending on type and expected activity concentration of the sample. Once a submenu is called the user is systematically taken through a series of operations to complete a task. In the vegetation laboratory, each sample is sectioned and placed in plastic cartons in preparation for freeze drying. Sample bar code labels are printed for each sample carton—the carton tare wets and wet/dry weights are all transferred electronically from balances into the LAB.DBF database. The use of sample bar code labeling and automation has totally eliminated transcriptions errors that previously plagued our sample processing and canning operations. Sample can weights (used for gamma-spectrometric measurements) and normalization weights are automatically computed and assigned to appropriate fields in the original SAMP.DBF database. The main menu also provides a user program submenu to help identify missing samples; a print submenu for bar code labels, chain of custody reports, and weekly summary reports; and a utilities submenu that contains the duplicate sample listings, separate box listings showing the number of blind duplicates and standards required in order to comply with quality control objectives, a sample locator menu to help with sample tracking and finally a updated storage transportainer listing showing the exact location of samples stored in our permanent archive. We have standardized on using the metal gamma cans as the sample archive. As analyses are completed, and the relevant Quality Control (QC) documentation generated and approved, the residual (leftover) sample material is disposed of in a certified waste stream.

Instrument Validation and Calibration

Our gamma-spectrometry facility houses a total of 24 high-resolution gamma detectors coupled to DEC VAXStation operating under Canberra/Nuclear Data systems data acquisition and reduction software. Each detector has been calibrated using natural matrix samples spiked with a mixed gamma solution traceable to the National Institute of Standards and Technology (NIST). To help maintain a consistency of measurement performance we have attempted to standardize on particular container (geometry considerations) and matrix types defined within the scope of our research program. In the past, we have also used commercial suppliers to prepare and calibrate our matrix calibration standards requiring that all products are traceable to NIST. We also maintain a 32 detector alpha-spectrometry system, two Gamma Products, Inc., Gas Proportional Alpha-Beta Counters, a Canberra Packard Liquid Scintillation Analyzer and a fully functional radiochemical laboratory. Radioactive tracers used for isotopic dilution and alpha spectrometry have all been independently certified by other laboratories. We regularly control and evaluate standard and blank analyses. All the Marshall Islands field, laboratory and assessment practices have been well documented [3–8] and/or appear in peer review literature[1–9], and conform with internationally accepted guidelines.[10–13]

Over 7,000 gamma-spectrometric measurements were performed as part of the Marshall Islands Program over the past 12 months. Consequently, much of the following discussion relates directly to our gamma-spectrometry facility. Similar quality control protocols and practices are applied in our radioanalytical studies but will not be discussed in detail.

The following laboratory practices have been included in our quality assurance plan for the gamma-spectrometry facility:

1. All calibrations should be performed using natural matrix standards traceable to NIST (or equivalent national standards).
2. Backgrounds and detector energy/efficiency calibrations shall be verified on each operable detectors on a weekly basis. Data will be evaluated and documented. Updated quality control charts (or listings) shall be maintained on FWHM characteristics, detector background/MDA's and detector efficiencies. An example output from the weekly efficiency and background validation counts is shown in Fig. 2 for detector OP during the period 1996-98.
3. The facility manager shall document/report any changes and/or occurrences.
4. Laboratory conditions including temperature, humidity and line voltage will be monitored on a continuous basis.

Also, as part of the gamma facilities internal quality control program we routinely perform duplicate counts on random samples. These duplicate counts are made on different detectors and are termed cross counts. Cross counts are used to provide assurances about the consistently of our measurements, and allow a more meaningful evaluation of actual duplicate analyses where homogeneity may influence the radionuclide content in different aliquots of the same sample. During the period between 1 January 1996 and 1 January 1998 a total of 567 cross count measurements were performed (Fig. 3.). The average recovery on duplicate counts for ^{137}Cs was 100.3 ± 0.1% with a median value of 100.0%. This data shows the high quality and consistency of our measurement capabilities supporting the Marshall Islands Program.

Method performance testing, validation, development and documentation

Generally our external Quality Control procedures for gamma-spectrometry are evaluated on the basis of a DCD (Delivery Control Document) listing containing 72 samples (including blind duplicates and blind standards). A minimum of 10% duplicates and 5% blind standards are analyzed as part of each DCD. Performance measures for gamma and alpha spectrometric studies has been previously developed by.[4] Standards require a 100% compliance at a level of ±10% of the reference value (Fig. 4a). Satisfactory performance on blind duplicates requires acceptability that 80% of duplicate pairs differ by less than ±20% within each DCD listing (Fig. 4b). Blind duplicates provide both a basis for measurement assurance and sample homogeneity testing. Currently we are using two natural matrix soils and two vegetation samples as internal reference standards. One of the soil standards (8207 soil) was distributed by the International Atomic Energy Agency (IAEA) in a worldwide intercomparison exercise and is now available as an IAEA Standard Reference Material (IAEA 367). We also use an aged natural matrix, spiked vegetation sample (9303 potato flake) originally prepared by NIST as a mixed gamma calibration standard.

A key component of our external quality control program was been our continued participation in national and international intercomparison exercises including those coordinated by IAEA and the DOE (e.g., MAPEP, Mixed Analyte Performance Evaluation Program). Data reported on the last two MAPEP intercomparison exercises are shown in Table 1 along with the reference values. The average measurement bias across five reported gamma-emitters radionuclides was -1.24% on MAPEP-97-S4 (soil) and -1.03% on MAPEP-97-W4 (water).

We have also participated in split-sample analyses, intercomparison and standard certification exercises with a large number of other agencies and institutions worldwide: Environmental Measurements Laboratory (EML), U.S.A.; GSF Institute, Germany; International Atomic Energy Agency (IAEA), Monaco.; International Atomic Energy Agency (IAEA), Austria; Marshall Islands Nationwide Survey, Government of the Marshall Islands; National Institute of Standards and Technology (NIST), U.S.A.; National Institute of Radiological Sciences, Japan; Oregon State University (OSU), U.S.A.; Pacific Northwest Laboratory (NPL), U.S.A.; Thermal Analytical Norcal (TMA), U.S.A.; University of South Carolina, U.S.A.; University of Washington, U.S.A.; and the Western Oregon State College (WOSC), U.S.A.

Scientific Peer Program Review

Over the past 18 years there has been nine independent peer reviews of the Marshall Islands Dose Assessment and Radioecology Program. The most recent reviews were conducted in 1993/94, by a 15 member panel of scientists convened by the United States National Academy of Sciences[14], and in 1995/96, by a 12 member panel of international scientist coordinated by the International Atomic Energy Agency in Vienna, Austria[15]. The IAEA review concluded that on the basis of the amount and quality of data and scientific information submitted for review, that: no further independent corroboration of the measurements and assessments of the radiological conditions at Bikini Atoll is necessary[15]. This conclusion was reached on the basis of the excellent quality control of the measurements and assessments; the regular participation of our laboratory in various intercomparison exercise, and the results of an independent sampling and analysis program conducted by IAEA. Despite the very limited nature of the IAEA sample collection and analysis program both the mean values and ranges of activity concentrations for ^{137}Cs, ^{90}Sr, $^{239+240}$Pu and ^{241}Am in selected foodstuffs measured by the IAEA were in excellent agreement with LLNL data collected over the past 20 years (Table 2). These extensive independent national and international program reviews have proved to be invaluable in documenting the quality and credibility of our measurements, and have provided independent endorsement of our recommended remedial strategies.

Summary

The Marshall Islands Radioassay Quality Assurance Program aims at providing reliably accurate and traceable measurement data in order to make informed decisions about radiological conditions on the atolls, and to evaluate the need for and consequences of control measures. The broad international acceptance of our data and assessments has largely been developed through our internal quality assurance/quality control programs, long-standing contributions to national and international intercomparison exercises, and periodic program peer review.

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Table 1. Mixed Analyte Performance Evaluation Program—Results.

Exercise	Isotopes	Units	LLNL reported value	Unc.	MAPEP grand mean	Std dev.	MAPEP acceptance range	MAPEP reference value	LLNL bias (%)
MAPEP-97-S4 Soil	Americium-241	Bq kg^{-1}	17.2	1.0	18.5	2.2	13.4 – 24.9	19.2	-10.4
	Cesium-137	Bq kg^{-1}	795.1	0.4	797.4	65.4	543.9 – 1010.1	777.0	2.3
	Cobalt-57	Bq kg^{-1}	490.7	0.4	488.7	48.1	347.0 – 644.5	495.8	-1.0
	Cobalt-60	Bq kg^{-1}	863.3	0.3	865.2	69.1	606.0 – 1125.5	865.8	-0.3
	Manganese-54	Bq kg^{-1}	207.7	1.3	214.7	20.1	145.0 – 269.3	207.2	0.2
	Plutonium-238	Bq kg^{-1}	21.4	0.8	26.5	2.7	18.8 – 34.9	26.9	-20.3
	Plutonium-239/240	Bq kg^{-1}	33.4	1.2	38.7	3.6	27.9 – 51.8	39.9	-16.3
	Potassium-40	Bq kg^{-1}	609.4	1.9	640.0	63.5	456.4 – 847.7	652.1	-6.6
	Zinc-65	Bq kg^{-1}	1732.8	0.5	1791.3	173.9	1191.4 – 2212.6	1702.0	1.8
MAPAP-97-W5 Water	Americium-241	Bq L^{-1}	1.91	0.02	2.0	0.2	1.49 – 2.77	2.1	-10.3
	Cesium-137	Bq L^{-1}	215.2	2.1	214.3	15.3	149.1 – 277.0	213.1	1.0
	Cobalt-57	Bq L^{-1}	270.9	1.7	270.1	18.1	194.2 – 360.7	277.5	-2.4
	Cobalt-60	Bq L^{-1}	131.5	1.3	131.0	8.3	92.7 – 172.2	132.5	-0.7
	Manganese-54	Bq L^{-1}	217	2.7	221.2	16.9	155.14 – 288.1	221.6	-2.1
	Plutonium-238	Bq L^{-1}	1.39	0.03	1.3	0.1	0.98 – 1.82	1.4	-0.7
	Plutonium-239/240	Bq L^{-1}	3.44	0.07	3.3	0.2	2.41 – 4.47	3.4	0.0
	Zinc-65	Bq L^{-1}	583	6.7	599.3	55.9	411.8 – 764.7	588.3	-0.9

Table 2. Corroborative measurements performed by IAEA on radionuclide concentrations in different foodstuffs collected from Bikini Island in 1997 (Bq g^{-1}, wet wt.).

Food type	Radionuclide	Mean	Minimum	Maximum
Coconut Fluid				
LLNL	^{137}Cs	1.2×10^{0}	2.8×10^{-2}	6.1×10^{0}
IAEA		8.0×10^{-1}	8.4×10^{-2}	2.4×10^{0}
LLNL	^{90}Sr	4.5×10^{-4}	7.1×10^{-5}	8.8×10^{-4}
IAEA		—	4.9×10^{-4}	6.3×10^{-3}
LLNL	$^{239+240}Pu$	1.0×10^{-6}	3.3×10^{-7}	2.2×10^{-6}
IAEA			2.0×10^{-7}	4.2×10^{-7}
LLNL[a]	^{241}Am	8.5×10^{-6}	—	—
IAEA		—	1.1×10^{-7}	$<3.4 \times 10^{-7}$
Coconut Meat				
LLNL	^{137}Cs	2.9×10^{0}	1.4×10^{-1}	6.1×10^{0}
IAEA		1.2×10^{0}	1.2×10^{-1}	3.8×10^{0}
LLNL	^{90}Sr	5.9×10^{-3}	6.3×10^{-4}	2.9×10^{-2}
IAEA		—	3.0×10^{-3}	1.5×10^{-2}
LLNL	$^{239+240}Pu$	2.7×10^{-6}	1.7×10^{-7}	1.4×10^{-5}
IAEA		—	$<2.7 \times 10^{-7}$	$<4.8 \times 10^{-7}$
LLNL	^{241}Am	3.610×10^{-6}	5.0×10^{-7}	1.6×10^{-5}
IAEA		—	$<4 \times 10^{-7}$	$<4.2 \times 10^{-7}$
Pandanus fruit				
LLNL	^{137}Cs	3.9×10^{0}	1.1×10^{-1}	1.9×10^{1}
IAEA		8.1×10^{0}	1.0×10^{0}	2.1×10^{1}
LLNL	^{90}Sr	1.2×10^{-1}	8.6×10^{-3}	3.6×10^{-1}
IAEA		—	1.5×10^{-2}	2.1×10^{-1}
LLNL	$^{239+240}Pu$	3.2×10^{-6}	3.4×10^{-7}	1.1×10^{-5}
IAEA		—	8.4×10^{-7}	3.6×10^{-4}
LLNL	^{241}Am	3.8×10^{-6}	2.8×10^{-7}	1.5×10^{-5}
IAEA		—	5.0×10^{-7}	1.7×10^{-5}
Breadfruit				—
LLNL	^{137}Cs	3.8×10^{-1}	9.0×10^{-2}	1.1×10^{0}
IAEA		1.5×10^{-1}	1.2×10^{-1}	1.7×10^{-1}
LLNL	^{90}Sr	6.9×10^{-2}	6.0×10^{-3}	2.0×10^{-1}
IAEA[a]		2.7×10^{-2}	—	—
LLNL	$^{239+240}Pu$	1.8×10^{-6}	1.4×10^{-7}	9.8×10^{-6}
IAEA[a]		1.1×10^{-7}	—	
LLNL	^{241}Am	1.2×10^{-6}	1.5×10^{-7}	5.3×10^{-6}
IAEA		$<3.7 \times 10^{-7}$	—	—

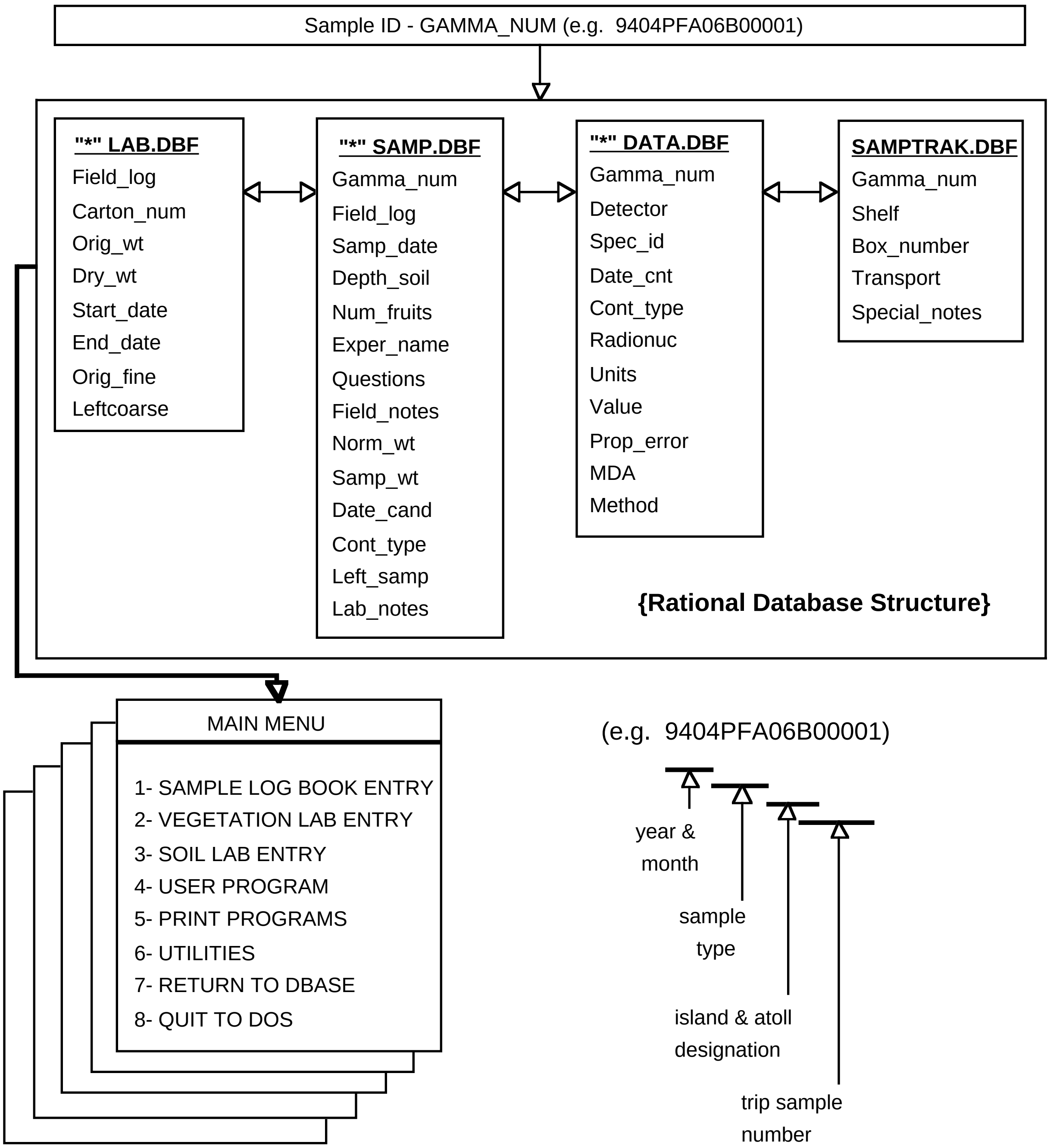
Source (IAEA, 1998); [a] Single values.

Figure 1. Marshall Islands database structure and data management profile.

Figure 2. Weekly instrument validation for the period 1996–98—detector OP at 661 keV.

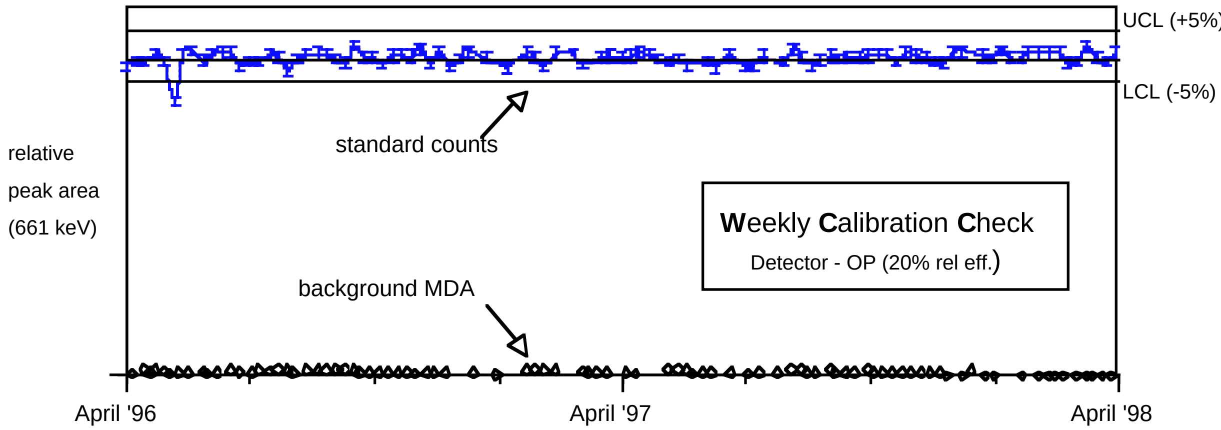
Figure 3. Internal quality control measurements—cross counts for ^{137}Cs for the period 1996–98.

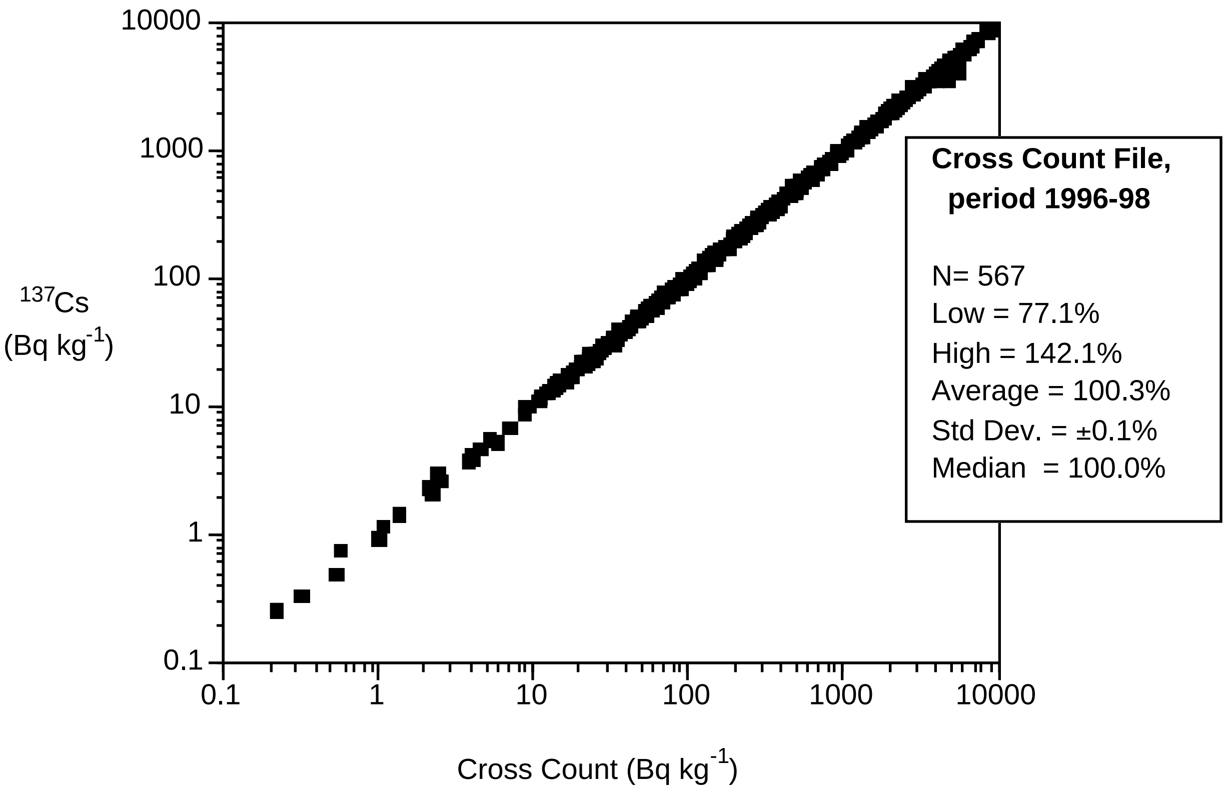
Figure 4. Internal Quality Control. Charts (A) Measurement of blind standards on internal reference standard LLNL 8510, and (B) Measurement of sample duplicates, for the period 1996–98.

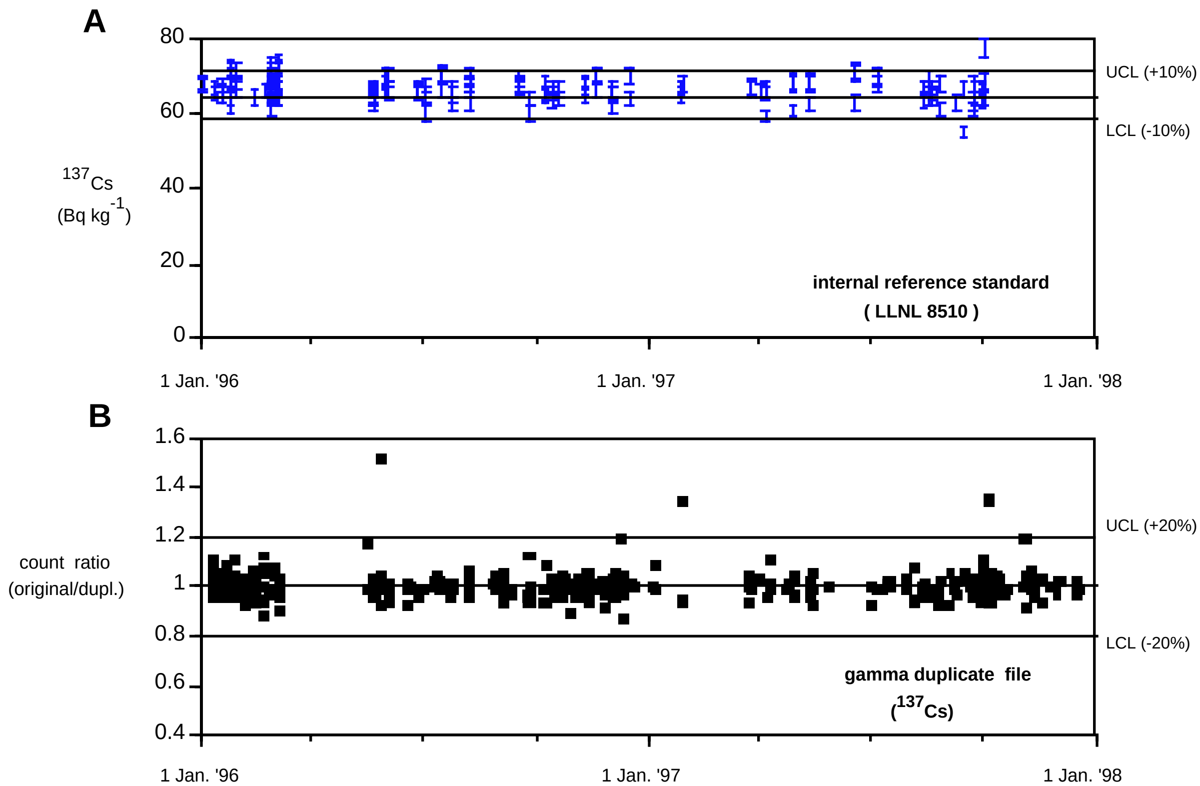


{Menu driven data entry screens}

{sample ID - GAMMA_NUM}


UCL (+5%)
LCL (-5%)
standard counts
relative
peak area
(661 keV)
Weekly Calibration Check
Detector - OP (20% rel eff.)
background MDA
April '96
April '97
April '98


10000
1000
100
10
1
0.1
137Cs
(Bq kg-1)
0.1
1
10
100
1000
10000
Cross Count (Bq kg-1)
Cross Count File,
period 1996-98
N= 567
Low = 77.1%
High = 142.1%
Average = 100.3%
Std Dev. = ±0.1%
Median = 100.0%


A
80
60
40
20
0
137Cs
(Bq kg-1)
UCL (+10%)
LCL (-10%)
internal reference standard
(LLNL 8510)
1 Jan. '96
1 Jan. '97
1 Jan. '98
B
1.6
1.4
1.2
1
0.8
0.6
0.4
count ratio
(original/dupl.)
UCL (+20%)
LCL (-20%)
gamma duplicate file
(137Cs)
1 Jan. '96
1 Jan. '97
1 Jan. '98